

Long Duration Energy Storage Using Hydrogen in Metal–Organic Frameworks: Opportunities and Challenges

Peng Peng, Henry Z. H. Jiang, Stephanie Collins, Hiroyasu Furukawa, Jeffrey R. Long, and Hanna Breunig*



Cite This: *ACS Energy Lett.* 2024, 9, 2727–2735



Read Online

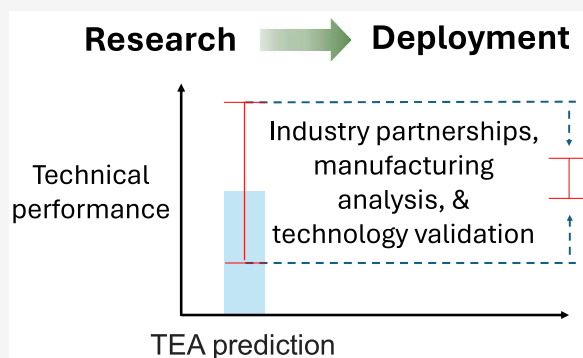
ACCESS |

Metrics & More

Article Recommendations

Supporting Information

ABSTRACT: Materials-based H₂ storage plays a critical role in facilitating H₂ as a low-carbon energy carrier, but there remains limited guidance on the technical performance necessary for specific applications. Metal–organic framework (MOF) adsorbents have shown potential in power applications, but need to demonstrate economic promises against incumbent compressed H₂ storage. Herein, we evaluate the potential impact of material properties, charge/discharge patterns, and propose targets for MOFs' deployment in long-duration energy storage applications including backup, load optimization, and hybrid power. We find that state-of-the-art MOF could outperform cryogenic storage and 350 bar compressed storage in applications requiring ≤ 8 cycles per year, but need ≥ 5 g/L increase in uptake to be cost-competitive for applications that require ≥ 30 cycles per year. Existing challenges include manufacturing at scale and quantifying the economic value of lower-pressure storage. Lastly, future research needs are identified including integrating thermodynamic effects and degradation mechanisms.



Hydrogen (H₂) is a promising energy storage material and is expected to play an important role in a decarbonized society.^{1,2} Successfully deploying a hydrogen-based economy by 2030 can reduce U.S. emissions by 16%, and create up to \$140 billion in revenues and 700,000 jobs.³ Developing solutions to affordably store H₂ for long duration is essential for coupling H₂ based on renewable intermittent energy with power applications that have little flexibility on time-of-use.⁴ Conventional H₂ storage methods mainly rely on high pressures (as high as 700 bar) or liquefaction to below boiling point (20 K) to achieve high densities.⁵ The compression processes and the associated tanks are costly, have large land footprints, pose safety concerns for high pressure, and liquid storage faces challenges with high boil-off rates that limit storage duration.^{6,7}

Presently, it is unclear how material-based storage systems perform compared to compressed gas and cryogenic liquid hydrogen storage for long-duration energy storage, and what are the targets for materials to outperform them on a cost basis. Chemical H₂ storage methods convert H₂ to a storage material with high hydrogen content, such as ammonia, liquid organic hydrogen carriers such as methanol or methylcyclohexane, and metal hydrides.^{8–10} Ammonia and carriers are emerging as

frontrunners for bulk H₂ transportation applications.^{11,12} However, they require careful thermal management to protect expensive catalysts, high temperatures to facilitate conversion, and complex purification, which may not scale or be responsive enough for power applications coupled with turbines or fuel cells.⁹

Physical storage methods rely on adsorbent materials to obtain a high H₂ storage capacity under mild pressure and temperature conditions.¹³ Hydrogen can be released at a tunable pressure (below 200 bar), and rapidly as H₂ is stored as a dihydrogen molecule without the need for chemical or catalytic processes.¹⁴ Among the adsorbents under consideration, metal–organic frameworks (MOFs) present a class of porous materials based upon metal ions connected via organic linkers. MOFs have been viewed as promising materials for H₂

Received: March 28, 2024

Revised: April 25, 2024

Accepted: May 1, 2024

Published: May 14, 2024



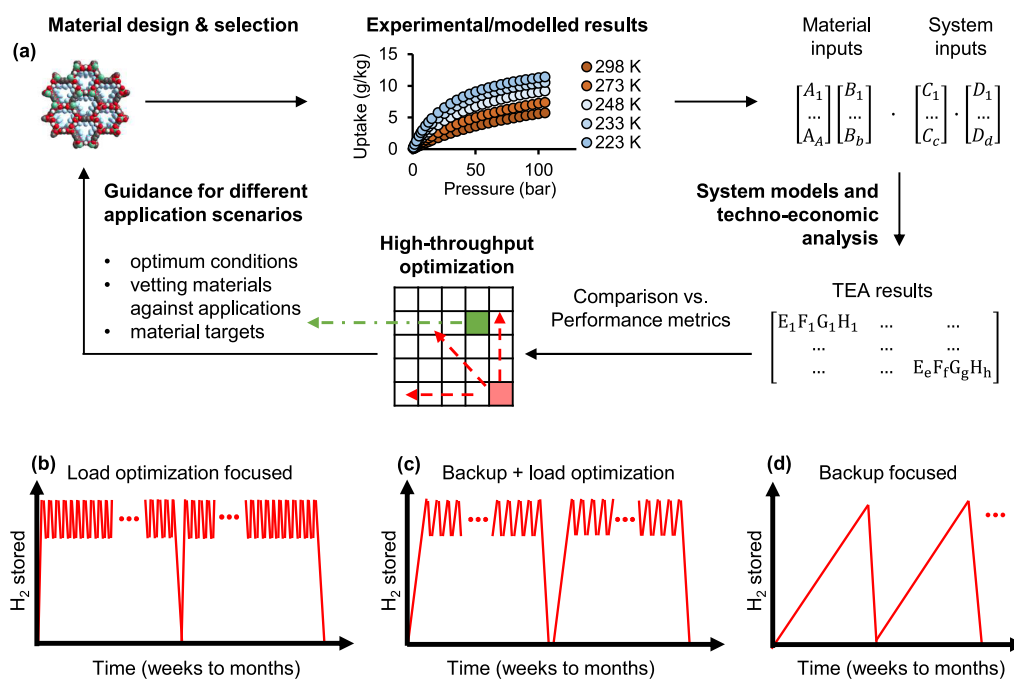


Figure 1. Overview of the scope of the study. (a). Proposed integration of systems analysis within material design and selection research. (b–d). Representative charge and discharge patterns for the H₂ storage system in prototypical long-duration energy storage applications.

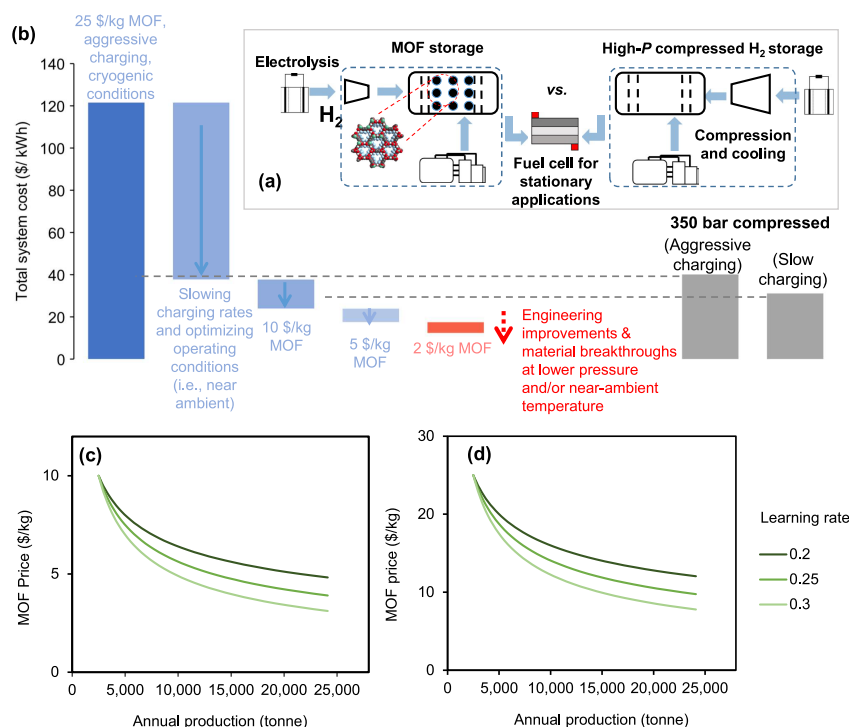


Figure 2. Summary of the status and research roadmap for MOFs to be comparable with compressed H₂ storage at 350 bar. (a). The schematic showing the key system components and their integration included in this study. (b). A roadmap for lowering capital costs of the H₂ storage system (total system cost) from a starting point of MOF-5 under 77 K and 170 bar, with the rest using Ni₂(*m*-dobdc) as representative state-of-the-art MOF. Further cost sensitivities can be found later in Figures 3 and 4. Aggressive charging and slow charging represent ~70 and 8 cycles of operation per year, respectively. (c)–(d). Projected cost of MOF under common learning rates from 0.2 to 0.3 under various initial conditions (c) 10 \$/kg, (d) 25 \$/kg, where annual production starts at 2500 tonnes, assuming growing at a reported rate of 12% per year.^{23–25}

storage due to their high surface area (over 1000 m²/g), and the high tunability of their chemical and structural properties, allowing them in some cases to bind hydrogen at open metal coordination sites.^{15,16} Because of this high tunability, there is

room for meeting challenging cost targets via synergistically designing system operation and tuning MOFs. In other words, if not comprehensively studied, a risk is run that a given MOF may be evaluated in techno-economic analyses (TEAs) under

operating conditions missing its optimized use, and thus not providing a definitive assessment of the technology's potential for deployment.

Considering the above challenges, and the need to accelerate the implementation of H₂ storage technologies, in this study we develop and apply an approach to identify ideal MOF properties based on target long-duration energy storage applications characterized by load profiles that represent backup power (8 or fewer cycles per year), load optimization, and hybrid applications requiring up to 30 cycles per year (Figure 1). We categorize existing MOFs into representative types based on the operating conditions and load profiles that minimize the overall levelized cost of storage (LCOS). The overall framework involves automated TEA based on materials-level inputs (e.g., skeletal density, H₂ uptake, and adsorption enthalpy), as well as system-level inputs such as tank design, insulation, and charging patterns for long-duration storage. As work was already performed demonstrating cryogenic liquid hydrogen storage is nonviable in long-duration energy storage applications (30 or fewer cycles per year) coupled with on-site H₂ generation, we focus on 350 bar above-ground compressed gas storage as a benchmark (Figure 2a), which is set as 31–40 \$/kWh installed capital cost.^{5,17} Note that other reported reference values include ~17 \$/kWh for above-ground storage tank only, and underground compressed storage at a similar scale.^{18,19}

Opportunities for capital cost savings include material development, optimized operating cycles, and lower material costs.

The potential for each opportunity to lower total system capital costs is summarized in Figure 2b. We find that for a given application and type of MOF, many combinations of variables and operating conditions can reach the same cost target and it is necessary to screen for the most viable designs (Supporting Information (SI) Section S2). The roadmap presents the total system cost for an energy storage system using MOF-5 at conditions where it has its highest H₂ uptake (dark blue column). For MOFs produced at 25 \$/kg, and operating under cryogenic and high-pressure conditions seen in experimental isotherms, the system cost is around three times as much as compressed gas at 350 bar if aiming for fast charging for both systems (70 cycles per year).¹⁴ Substantial cost reductions (~67% reduction from ~120 \$/kWh to ~40 \$/kWh) are possible by moving to materials with high uptake at near ambient temperatures such as Ni₂(*m*-dobdc), by optimizing storage pressure and temperature, and by relaxing requirements on fill rates (8 cycles per year). Developing materials with improvements in excess uptake can be achieved by either improving the density of adsorbent sites per volume or mass of material, or improving the efficiency of those sites given a set of adsorption/desorption conditions by tuning thermodynamics.¹⁶

Given the challenge of material discovery and validation, alternatively, combining system-level analyses with engineering improvements, such as lowering the cost of storage tanks and refrigeration systems, and improved heat integration, could enable MOF systems to become the leading physical H₂ storage option for long-duration energy storage. In the near term, known improvements in material manufacturing and

improved system integration to properly set charging patterns will lower the deployment cost of known materials (details on charging patterns are in SI Section S3). As noted in Figure 2b, by reducing the MOF manufacturing cost to the proposed lowest point of 10 \$/kg reported in the literature,^{20,21} MOF-H₂ storage under slow charging could start to be comparable with compressed 350 H₂ storage.

Recent studies project MOF manufacturing costs between 10 \$/kg and 70 \$/kg, depending on the synthetic route (e.g., thermal-based, solvent-based, production from waste).^{20–22} A MOF manufacturing cost of 10 \$/kg reflects production scales of 2500 tonnes per year.²⁰ In comparison, a single stationary 10 MW, 96-h storage system requires around the level of thousands of tonnes of MOFs depending on operating conditions. Increasing production at the demonstrated rate of 12%²³ could lead to costs as low as 5–10 \$/kg (Figure 2c and 2d), if the learning rate of MOF is between 0.2 and 0.3, which is a common range for emerging technologies and has been used for predicting the cost of H₂ technologies.^{24,25} Achieving sub-5 \$/kg would likely be challenging for any MOFs that rely on critical materials, and require a faster ramp-up in production volume (e.g., 30% as shown in SI Figure S1).

Moving on to consider both operating and capital expenses for a storage system is possibly by determining the LCOS. The LCOS captures additional fixed and variable operating costs (related assumptions such as operating pattern and insulation can be found in SI Sections S1 to S3). The LCOS for a given application can vary significantly with the selection of storage tank operating temperatures and pressures due to trade-offs in H₂ uptake and H₂ conditioning (compression and cooling) costs. Such variations are nonintuitive, and necessitate optimization to identify the best performance for a single MOF under practical ranges in storage tank pressure and temperatures. For example, in Figure 3a, the lowest LCOS for Ni₂(*m*-dobdc) under a moderate charging rate (50% of discharge) is 0.17 \$/kWh under 208 K and 86 bar (region 1). The breakdown of the LCOS is presented in Figure 3b highlighting the importance of both capital and operating costs. Under this scenario, the refrigeration and compressor cost is comparable with the storage tank cost (Figure 3b), whereas, under the aggressive charging and/or colder conditions used in previous work, refrigeration costs dominate.¹⁴

However, in some cases, there may be little control over such factors, and it is therefore illustrative to identify the necessary improvement in excess H₂ uptake to be cost-competitive with a compressed gas system (SI Section S2). By applying this strategy to a scenario where the storage tank is cycled 30 times per year, we find that Ni₂(*m*-dobdc) can outperform 350 bar compressed gas storage in LCOS under slow charging conditions at 5 \$/kg MOF, but needs an increased uptake (>5 g/L) under fast charging scenarios (SI Figure S2).

Further, we show that for Ni₂(*m*-dobdc), the uncertainties of porosity and manufacturing cost significantly influence the optimal storage tank operating pressure (Figure 3c; input range determined in SI Section S2). For example, low porosity favors mild cooling conditions (~200 K) to increase uptake. For more expensive MOFs, higher pressure and lower temperatures are favored to maximize energy density and reduce sorbent usage in general. Therefore, when MOF price is around 15 \$/kg or above, the optimum conditions are found under 170 bar (highest value modeled as described in SI

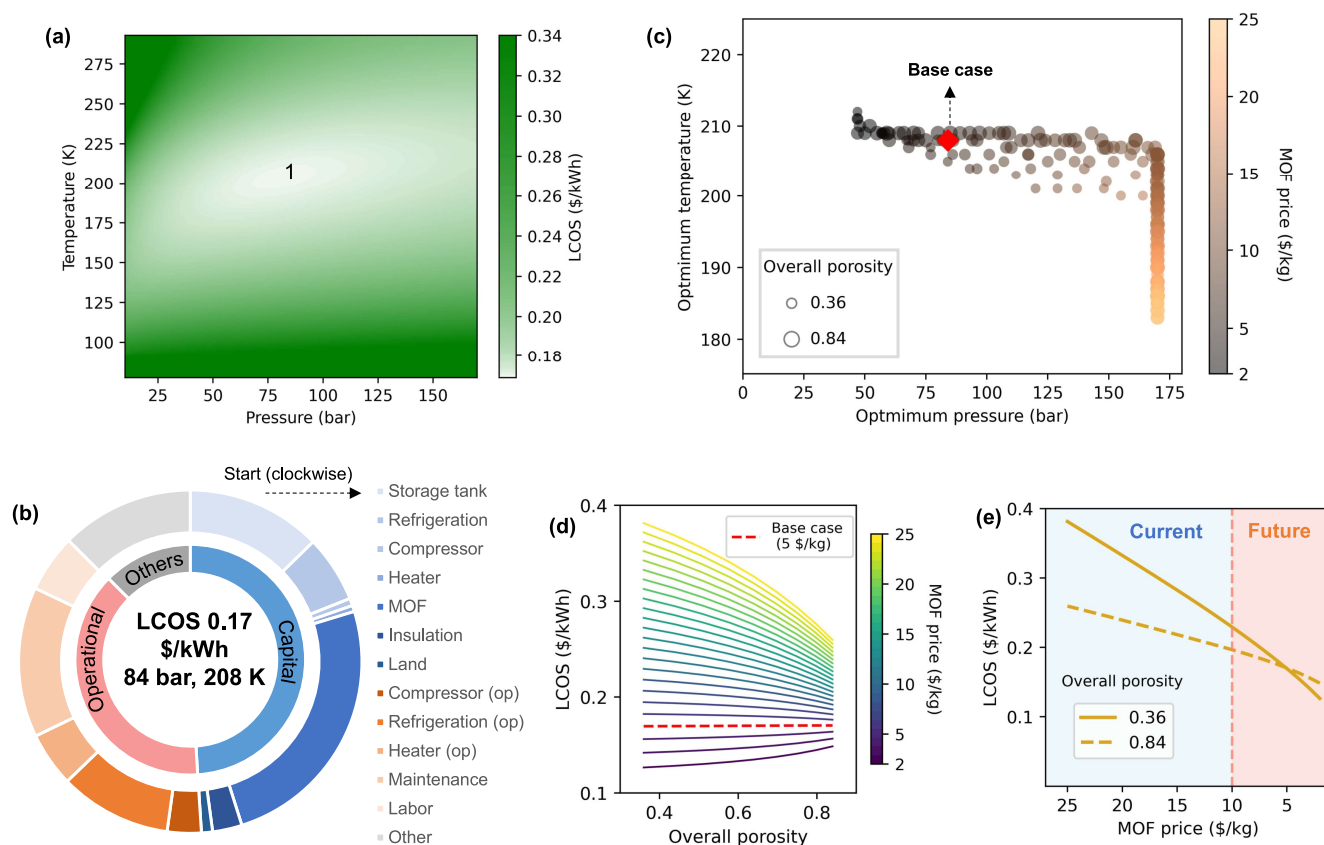


Figure 3. Optimization and uncertainties of MOF performance. (a) LCOS performance profile for $\text{Ni}_2(m\text{-dobdc})$ at 5 \$/kg, overall porosity = 0.64 (bed and pellet porosities = 0.4), charge rate half as fast as discharge rate. Region 1 denotes the conditions with optimal LCOS for this application. (b) Breakdown of the lowest LCOS for $\text{Ni}_2(m\text{-dobdc})$. (c) Ranges of optimum conditions for $\text{Ni}_2(m\text{-dobdc})$ MOF under different assumptions of overall porosity (pellet and bed) from 0.36 to 0.84, cost of manufacturing from 2 \$/kg to 25 \$/kg (base case 0.64 overall porosity, 5 \$/kg MOF). (d) and (e) Effects of MOF manufacturing cost and overall porosity on the LCOS performance of $\text{Ni}_2(m\text{-dobdc})$.

The challenge of applying a techno-economic analysis to inform prototype design and storage technology evaluation is that there are many uncertainties associated with the scaled-up performance of MOFs that require further demonstration.

Section S1). These factors collectively can also have a large influence on LCOS (Figures 3d and 3e).

Another challenge related to the development of MOFs for long-duration energy storage is the magnitude of plausible materials requiring consideration due to MOF tunability. Thus, further, we show how to evaluate specific MOFs in various application settings. In the process, three types of MOFs have emerged (proposed in Table 1), described below. In the final portion of this perspective, we explore whether certain classes of MOFs are preferred candidates for specific long-duration energy storage markets under low and high electricity and land costs.

To date, most MOFs studied for H_2 storage are Type 1, including MOF-5, HKUST-1, UiO-67, ZIF-8, MIL-100,

Table 1. Representative Types of MOFs Investigated for H_2 Storage

	MOF types		
	Type 1	Type 2	Type 3
Representative MOFs	MOF-5, HKUST-1, UiO-67, ZIF-8, MIL-100, etc.	$\text{Ni}_2(m\text{-dobdc})$, V-btdd	ALF
Target operation conditions	77K	Relatively closer to ambient	Subambient and low pressure
Main desirable nature of MOFs	High surface area	Open metal sites	Small pores and designed to use economical raw materials (i.e., abundant metals and linkers)
Current status	Abundant experiments and models available (77 K, 5–200 bar)	Moderate experiments and models available	Limited experiments and models available
Refs	2,26–30	31,32	33

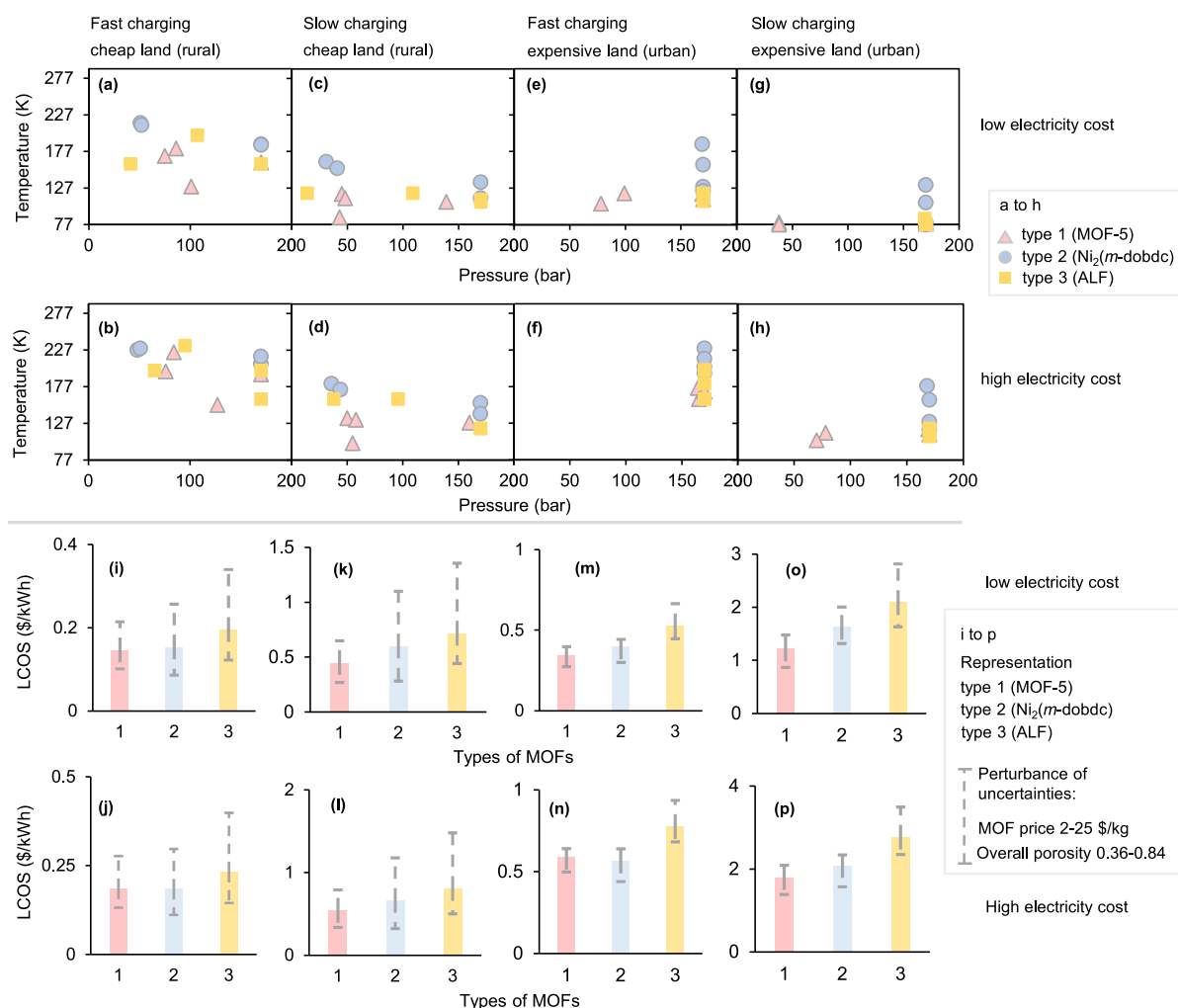


Figure 4. Summary of how MOF types and application deployment conditions could affect the optimum storage tank conditions and LCOS. (a) to (h) potential changes to optimum tank pressure and temperature for three types of representative MOFs under different market conditions. (i) to (p) potential changes to LCOS for three types of representative MOFs under the market conditions represented in (a) to (h). The error bar for each MOF type represents perturbation of low and high pellet and bed porosities of 0.2 and 0.6 (representing overall porosities of 0.36 and 0.84), with low and high material costs of 2 \$/kg and 25 \$/kg.

etc.^{26–30,34} They are characterized as having decent uptake under cryogenic conditions such as 77 K, and tend to favor cooled operation (SI Section S5), which is also the range that is mostly studied for these MOFs. Note performance trends are the most generalizable for Type 1 MOFs since abundant data from multiple similar MOFs (noticeably higher uptake under cryogenic conditions) is used to develop the trend, as shown in SI Section S5.

Type 2 MOFs include $\text{Ni}_2(m\text{-dobdc})$ and V-btdd, and have higher uptake compared with Type 1 MOFs at near-ambient temperatures,^{31,32} therefore requiring less cooling than Type 1 to reach the required storage capacity, for example under conditions (170–230 K depending on MOF type and application patterns). The success of $\text{Ni}_2(m\text{-dobdc})$ and V-btdd achieving record-high H_2 uptake under relatively closer to ambient conditions is partially due to the fact that their enthalpy changes during adsorption (ΔH) are close to, or within the optimum range of -15 to -25 kJ/mol, while showing high density of open metal site and relatively small pore diameter.^{31,32,35–37} Compared with prototypical Type 1 MOFs such as MOF-5, HKUST-1, UiO-67, ZIF-8, and MIL-

100, Type 2 MOFs open up new opportunities as they circumvent the need for excessive cooling (SI Section S5).

Recently, a third type of MOF has emerged in the H_2 storage literature, which is designed to be synthesized via abundant and cheap materials (i.e., aluminum and formic acid), while retaining H_2 uptake. Type 3 MOF requires more cooling than Type 2 but reaches its peak performance under low-pressure conditions.³³ Evans et al. report a low optimum pressure range (10–40 bar), which remains high enough to transfer H_2 into fuel cells.³³ The generalization for Type 2 and Type 3 MOFs needs further evaluation, which requires more experimental data, particularly for Type 3, related to isotherm assumptions under broad temperature ranges (see SI Section S2 for discussion).

From here, we identify the optimum operating conditions for these materials under different application scenarios (Figures 4a to h) to predict their best cost performance (Figures 4i to p), accounting for uncertainties in materials properties. The application scenarios are primarily governed by the high and low bounds for land cost, ratio of charge and discharge rates, and electricity costs (SI Sections S1 and S6). Land cost can have significant implications on system design,

favoring densification, but only for very costly locations such as metropolitan regions (SI Section S6). When the land price is high, high pressures are favored for all three types to maximize energy density, and optimum temperature is lower when slow charging is allowed as refrigeration becomes less of a concern. Of all scenarios modeled, Type 2 MOFs, represented by $\text{Ni}_2(m\text{-dobdc})$, offer the lowest cost if they can be produced cheaply. In comparison, Type 1 MOFs are more sensitive to application settings, where they manage to achieve a lower cost than Type 2 by a small amount under certain scenarios. Despite having higher LCOS under the same MOF price range, Type 3 MOFs have higher chances of achieving the cheapest material cost using abundant metals and linkers. Further, their capability of achieving moderate uptake under low pressure (~ 10 g/kg at ~ 10 bar) could make them an ideal candidate for high-pressure electrolyzers (>30 bar, while this Perspective assumes 2 bar).^{33,38}

Besides pressure, heat and cooling management is affected by the way in which storage systems are integrated into H_2 production and end use. When coupled with electrolyzers, MOF-based H_2 storage requires that the H_2 be compressed and cooled at the same time, which increases energy consumption. For reference, the energy consumption to store electrolysis-produced H_2 in $\text{Ni}_2(m\text{-dobdc})$ system under optimal base case (84 bar, 208 K) is ~ 6.8 kWh/kg, which is comparable to the energy required for 350 bar compressed storage ~ 9.3 kWh/kg in this study (include intercooling to ambient conditions). On the other hand, using effective cooling management and advanced refrigeration systems such as mix-cycle and cascade refrigeration could potentially reduce energy consumption for H_2 liquefaction by up to 50%.^{39,40} Such experience would be beneficial to optimizing MOF-based H_2 storage systems to improve energy efficiency and reduce cost.

As suggested in this Perspective, the uncertainty regarding optimal tank operation and ultimate LCOS is large and will remain until demonstrations can yield new insights on parameters such as material stability, material thermodynamic properties and heat management, packing porosities in tanks and manufacturing pathways, and the end user flexibility regarding use cycles. Material stability and reducing degradation are moderately important factors for the success of MOFs in energy storage applications with low cycling and long IDLE time. Establishing longer cycle stability (i.e., thousands of cycles) and higher tolerance to impurities could open more opportunities in H_2 storage applications (such as peak shaving applications that require daily cycles). Additionally, the insights on heat management gleaned from a prototype or small pilot demonstration may not translate well to larger scales where novel system integrations are possible. Although MOFs generally experience low thermal conductivities⁴¹ that could limit rates of heat transfer, this may not be a limiting factor in slow-charging applications. Still, correctly measuring thermodynamic properties and understanding the impacts of heat transfer rates will be important for conducting more detailed engineering analyses of heating/cooling management techniques, particularly those that reduce cooling loads and avoid the use of natural gas for heat.

Fundamental research remains critical for advancing MOFs in H_2 storage. First, the thermodynamic properties of MOFs are tunable, and the combined effects between changing standard enthalpy (ΔH°) and entropy (ΔS°) upon adsorption govern the equilibrium capacities of H_2 during adsorption and

As a bridge between bench and commercial-scale applications, kilogram to pilot-scale demonstrations could provide useful data and experience to address the above gaps. Given the importance of hydrogen storage for decarbonization, it is now time for fundamental research, engineering, industry, and analysis teams to work together and form more practical insights for deployment of MOFs in long-duration hydrogen storage.

desorption (SI Section S4). The goal being to yield a material with the largest percentage of working hydrogen binding sites between adsorption and desorption cycles (SI Figure S5), increases its chances of reaching the target uptake capacity, if not already met.^{42,43} This goal must be considered alongside the resulting changes to energy consumption at the system level (SI Figure S5e and S5f). Second, the correlation of porosity and surface area with system-level performance is still poorly understood, as it must be modeled via molecular simulations (such as Grand Canonical Monte Carlo simulations), or measured experimentally, which are computation or labor-expensive. Machine learning methods could greatly help with reducing computational speed and improving the optimization efficiency of performance prediction. As recent machine learning studies are focused on operating conditions around 77 K (Table 1),^{29,30} we encourage future computational work to include more near-ambient conditions suggested in this Perspective.

Lastly, there are social and environmental benefits of operating the sorbent system at lower pressure and operating it autonomously on stored H_2 rather than natural gas which have yet to be quantified. Currently, there is little understanding of the economic value of reduced system complexity (i.e., dispensing systems), decreased hydrogen leakages, or the enhanced safety associated with storing H_2 under lower pressures compared to 350 and 700 bar for stationary applications. One study on stationary refueling stations suggests that dispenser costs for high-pressure storage could increase costs by 10%.⁴⁴ We expect that there are additional costs for safety handling, leakage prevention throughout the dispense systems, degradation, and additional chances of failure associated with storing under high pressure. In addition, life cycle impact factors such as critical material usage, resource availability, ecotoxicity, end-of-life pathways, and greenhouse gas emissions during manufacturing are crucial, and would further distinguish the potential between MOFs with other toxic carriers, or within MOFs themselves. These factors are important for the deployment of H_2 storage technologies and should be considered for future studies, possibly using analysis techniques including Life Cycle Assessment (LCA) and Failure Mode, and Effects Analysis (FMEA), which could be applied to improve our understanding of the potential of MOFs for deployment in long-duration stationary applications.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsenergylett.4c00894>.

Methods, operation cycle, potential effects of thermodynamics, representation of other prototypical MOFs, compressed H₂ storage under different pressures, effects of land costs, and benefits of storing under lower pressures (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Hanna Breunig – Energy Analysis and Environmental Impacts Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; orcid.org/0000-0002-4727-424X; Email: hannabreunig@lbl.gov

Authors

Peng Peng – Energy Analysis and Environmental Impacts Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; orcid.org/0000-0001-6554-2247

Henry Z. H. Jiang – Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; Department of Chemistry, University of California, Berkeley, California 94720, United States

Stephanie Collins – Energy Analysis and Environmental Impacts Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; Department of Civil and Environmental Engineering, University of California, Berkeley, California 94720, United States

Hiroyasu Furukawa – Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; Department of Chemistry, University of California, Berkeley, California 94720, United States; orcid.org/0000-0002-6082-1738

Jeffrey R. Long – Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; Department of Chemistry, University of California, Berkeley, California 94720, United States; Department of Chemical and Biomolecular Engineering, University of California, Berkeley, California 94720, United States; orcid.org/0000-0002-5324-1321

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsenergylett.4c00894>

Notes

The authors declare the following competing financial interest(s): The University of California, Berkeley, has been issued a patent relating to the use of Ni₂(*m*-dobdc) on which J.R.L. is listed as a co-inventor and has applied for a patent relating to the use of V-btdd on which J.R.L. is listed as a co-inventor. The remaining authors declare no competing interests.

Biographies

Peng Peng is a Research Scientist and licensed Professional Engineer (P.E.) at Lawrence Berkeley National Laboratory. His research focuses on deploying energy storage and advanced manufacturing technologies through co-development of materials and systems, and improvements in economics and circularity.

Henry Z. H. Jiang is a Ph.D. candidate in the Long group at UC Berkeley. He received his B.Sci in Chemistry from the University of Sydney, Australia. His research is focused on investigating adsorption in metal–organic frameworks with in situ spectroscopy.

Stephanie Collins is a PhD student in Civil and Environmental Engineering at UC Berkeley. She received her M.S. in Chemistry from UC Berkeley and B.S. in Chemistry from Case Western Reserve University. Her research interests are energy storage systems, with emphasis on the life cycle environmental and economic impacts of renewable hydrogen.

Hiroyasu Furukawa is a project scientist in the Department of Chemistry at the University of California, Berkeley. He received his Ph.D. from the University of Tokyo. His main research interests are in the development of crystalline porous materials, including metal–organic frameworks, for the storage and separations of industrially important gases.

Jeffrey R. Long is the C. Judson King Distinguished Professor of Chemistry at the University of California, Berkeley and a Faculty Senior Scientist at Lawrence Berkeley National Laboratory. His research focuses on the development of new materials for potential applications in energy and sustainability. Webpage: <http://alchemy.cchem.berkeley.edu>

Hanna Breunig is a Staff Scientist and Deputy-Head of the Sustainable Energy and Environmental Systems Department at Lawrence Berkeley National Laboratory. Her group focuses on process design, modeling, and systems analysis of low Technology Readiness Level energy systems including hydrogen, energy storage, and negative emissions technologies. Webpage: <https://eta.lbl.gov/people/hanna-breunig>

■ ACKNOWLEDGMENTS

The authors gratefully acknowledge support from the Hydrogen Materials—Advanced Research Consortium (HyMARC) established as part of the Energy Materials Network under the US DOE Office of Energy Efficiency and Renewable Energy (EERE), Hydrogen and Fuel Cell Technologies Office, under contract number DE-AC02-05CH11231 with Lawrence Berkeley National Laboratory. The authors would like to thank Zeric Hulvey, Ned Stetson, and Marika Wieliczko from DOE for their insights. We thank Huayang Cui of Databricks Inc. for insights on automation and data handling. The United States Government retains, and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

■ REFERENCES

- (1) Majumdar, A.; Deutch, J. M.; Prasher, R. S.; Griffin, T. P. A framework for a hydrogen economy. *Joule* **2021**, *5* (8), 1905–1908.
- (2) Wu, S.; Salmon, N.; Li, M. M.-J.; Bañares-Alcántara, R.; Tsang, S. C. E. Energy decarbonization via green H₂ or NH₃? *ACS Energy Letters* **2022**, *7* (3), 1021–1033.
- (3) Lebrohui, B. E.; Djoupo, J. J.; Lamrani, B.; Benabdelaziz, K.; Kouksou, T. Global hydrogen development-A technological and geopolitical overview. *Int. J. Hydrogen Energy* **2022**, *47* (11), 7016–7048.
- (4) Albertus, P.; Manser, J. S.; Litzelman, S. Long-duration electricity storage applications, economics, and technologies. *Joule* **2020**, *4* (1), 21–32.

- (5) Yartys, V. A.; Lototsky, M. V. An overview of hydrogen storage methods. *Hydrogen materials science and chemistry of carbon nanomaterials*; Springer: 2005; pp 75–104.
- (6) Zhang, T.; Uratani, J.; Huang, Y.; Xu, L.; Griffiths, S.; Ding, Y. Hydrogen liquefaction and storage: Recent progress and perspectives. *Renewable and Sustainable Energy Reviews* **2023**, *176*, 113204.
- (7) Wieliczko, M.; Stetson, N. Hydrogen technologies for energy storage: A perspective. *MRS Energy & Sustainability* **2020**, *7*, No. E41.
- (8) MacFarlane, D. R.; Cherepanov, P. V.; Choi, J.; Suryanto, B. H. R.; Hodgetts, R. Y.; Bakker, J. M.; Vallana, F. M. F.; Simonov, A. N. A roadmap to the ammonia economy. *Joule* **2020**, *4* (6), 1186–1205.
- (9) Allendorf, M. D.; Stavila, V.; Snider, J. L.; Witman, M.; Bowden, M. E.; Brooks, K.; Tran, B. L.; Autrey, T. Challenges to developing materials for the transport and storage of hydrogen. *Nat. Chem.* **2022**, *14* (11), 1214–1223.
- (10) Peng, P.; Su, J.; Breunig, H. Benchmarking plasma and electrolysis decomposition technologies for ammonia to power generation. *Energy Conversion and Management* **2023**, *288*, 117166.
- (11) Nagashima, M. *Japan's hydrogen strategy and its economic and geopolitical implications*; Ifri: Paris, France, 2018.
- (12) Niermann, M.; Drünert, S.; Kaltschmitt, M.; Bonhoff, K. Liquid organic hydrogen carriers (LOHCs)-techno-economic analysis of LOHCs in a defined process chain. *Energy Environ. Sci.* **2019**, *12* (1), 290–307.
- (13) Nazir, G.; Rehman, A.; Hussain, S.; Aftab, S.; Heo, K.; Ikram, M.; Patil, S. A.; Aizaz Ud Din, M. Recent Advances and Reliable Assessment of Solid-State Materials for Hydrogen Storage: A Step Forward toward a Sustainable H₂ Economy. *Advanced Sustainable Systems* **2022**, *6* (11), 2200276.
- (14) Peng, P.; Anastasopoulou, A.; Brooks, K.; Furukawa, H.; Bowden, M. E.; Long, J. R.; Autrey, T.; Breunig, H. Cost and potential of metal-organic frameworks for hydrogen back-up power supply. *Nature Energy* **2022**, *7* (5), 448–458.
- (15) Chen, Z.; Kirlikovali, K. O.; Li, P.; Farha, O. K. Reticular chemistry for highly porous metal-organic frameworks: The chemistry and applications. *Accounts of chemical research* **2022**, *55* (4), 579–591.
- (16) Allendorf, M. D.; Hulvey, Z.; Gennett, T.; Ahmed, A.; Autrey, T.; Camp, J.; Cho, E. S.; Furukawa, H.; Haranczyk, M.; Head-Gordon, M.; et al. An assessment of strategies for the development of solid-state adsorbents for vehicular hydrogen storage. *Energy Environ. Sci.* **2018**, *11* (10), 2784–2812.
- (17) Department of Energy Hydrogen Program Plan. US Department of Energy, 2020. <https://www.hydrogen.energy.gov/docs/hydrogenprogramlibraries/pdfs/hydrogen-program-plan-2020.pdf> (accessed 11/16/2023).
- (18) Papadias, D. D.; Ahluwalia, R. K. Bulk storage of hydrogen. *Int. J. Hydrogen Energy* **2021**, *46* (70), 34527–34541.
- (19) Houchins, C.; James, B. D.; Acevedo, Y.; Watts, Z. *Final Report: Hydrogen Storage System Cost Analysis (2017–2021)*; Strategic Analysis, Inc.: 2022.
- (20) DeSantis, D.; Mason, J. A.; James, B. D.; Houchins, C.; Long, J. R.; Veenstra, M. Techno-economic analysis of metal-organic frameworks for hydrogen and natural gas storage. *Energy Fuels* **2017**, *31* (2), 2024–2032.
- (21) El-Sayed, E.-S. M.; Yuan, D. Waste to MOFs: sustainable linker, metal, and solvent sources for value-added MOF synthesis and applications. *Green Chem.* **2020**, *22* (13), 4082–4104.
- (22) Severino, M. I.; Gkaniatsou, E.; Nouar, F.; Pinto, M. L.; Serre, C. MOFs industrialization: a complete assessment of production costs. *Faraday Discuss.* **2021**, *231*, 326–341.
- (23) Chakraborty, D.; Yurdusen, A.; Mouchaham, G.; Nouar, F.; Serre, C. J. A. F. M. Large-Scale Production of Metal-Organic Frameworks. *Adv. Funct. Mater.* **2023**, 2309089.
- (24) Smith, S. J. Careful currency conversion. *Nature Energy* **2020**, *5* (1), 14–15.
- (25) Lee, B.; Lee, H.; Cho, H.-S.; Cho, W.-C.; Kim, C.-H.; Lim, H. Projected economic outlook and scenario analysis for H₂ production by alkaline water electrolysis on the basis of the unit electricity price, the learning rate, and the automation level. *Sustainable Energy & Fuels* **2019**, *3* (7), 1799–1807.
- (26) Ahmed, A.; Seth, S.; Purewal, J.; Wong-Foy, A. G.; Veenstra, M.; Matzger, A. J.; Siegel, D. J. Exceptional hydrogen storage achieved by screening nearly half a million metal-organic frameworks. *Nat. Commun.* **2019**, *10* (1), 1568.
- (27) Gómez-Gualdrón, D. A.; Colón, Y. J.; Zhang, X.; Wang, T. C.; Chen, Y.-S.; Hupp, J. T.; Yildirim, T.; Farha, O. K.; Zhang, J.; Snurr, R. Q. Evaluating topologically diverse metal-organic frameworks for cryo-adsorbed hydrogen storage. *Energy Environ. Sci.* **2016**, *9* (10), 3279–3289.
- (28) Suresh, K.; Aulakh, D.; Purewal, J.; Siegel, D. J.; Veenstra, M.; Matzger, A. J. Optimizing hydrogen storage in MOFs through engineering of crystal morphology and control of crystal size. *J. Am. Chem. Soc.* **2021**, *143* (28), 10727–10734.
- (29) Ahmed, A.; Siegel, D. J. Predicting hydrogen storage in MOFs via machine learning. *Patterns* **2021**, *2* (7), 100291.
- (30) Bobbitt, N. S.; Snurr, R. Q. Molecular modelling and machine learning for high-throughput screening of metal-organic frameworks for hydrogen storage. *Mol. Simul.* **2019**, *45* (14–15), 1069–1081.
- (31) Kapelewski, M. T.; Runcevski, T.; Tarver, J. D.; Jiang, H. Z. H.; Hurst, K. E.; Parilla, P. A.; Ayala, A.; Gennett, T.; FitzGerald, S. A.; Brown, C. M.; Long, J. R. Record high hydrogen storage capacity in the metal-organic framework Ni₂(m-dobdc) at near-ambient temperatures. *Chemistry of materials* **2018**, *30* (22), 8179–8189.
- (32) Jaramillo, D. E.; Jiang, H. Z. H.; Evans, H. A.; Chakraborty, R.; Furukawa, H.; Brown, C. M.; Head-Gordon, M.; Long, J. R. Ambient-temperature hydrogen storage via vanadium (II)-dihydrogen complexation in a metal-organic framework. *J. Am. Chem. Soc.* **2021**, *143* (16), 6248–6256.
- (33) Evans, H.; Yildirim, T.; Peng, P.; Cheng, Y.; Deng, Z.; Zhang, Q.; Mullangi, D.; Zhao, D.; Canepa, P.; Bruenig, H.; et al. Hydrogen Storage with Aluminum Formate, ALF: Experimental, Computational and Technoeconomic Studies. *J. Am. Chem. Soc.* **2023**, *145* (40), 22150–22157.
- (34) Glante, S.; Fischer, M.; Hartmann, M. Investigation of the optimum conditions for adsorptive hydrogen storage. *Emergent materials* **2021**, *4* (5), 1295–1303.
- (35) Arean, C. O.; Bonelli, B.; Delgado, M. R.; Garrone, E. Hydrogen storage via physisorption: the combined role of adsorption enthalpy and entropy. *Turkish Journal of Chemistry* **2009**, *33* (5), 599–606.
- (36) Bhatia, S. K.; Myers, A. L. Optimum conditions for adsorptive storage. *Langmuir* **2006**, *22* (4), 1688–1700.
- (37) Getman, R. B.; Bae, Y.-S.; Wilmer, C. E.; Snurr, R. Q. Review and analysis of molecular simulations of methane, hydrogen, and acetylene storage in metal-organic frameworks. *Chem. Rev.* **2012**, *112* (2), 703–723.
- (38) Salehmin, M. N. I.; Husaini, T.; Goh, J.; Sulong, A. B. J. E. C. Management, High-pressure PEM water electrolyser: A review on challenges and mitigation strategies towards green and low-cost hydrogen production. *Energy Conversion and Management* **2022**, *268*, 115985.
- (39) Al Ghafri, S. Z. S.; Munro, S.; Cardella, U.; Funke, T.; Notardonato, W.; Trusler, J. P. M.; Leachman, J.; Span, R.; Kamiya, S.; Pearce, G.; et al. Hydrogen liquefaction: a review of the fundamental physics, engineering practice and future opportunities. *Energy Environ. Sci.* **2022**, *15* (7), 2690–2731.
- (40) Abidin, Z.; Tang, C.; Liu, Y.; Catchpole, K. Large-scale stationary hydrogen storage via liquid organic hydrogen carriers. *IScience* **2021**, *24* (9), 102966.
- (41) Islamov, M.; Babaei, H.; Anderson, R.; Sezginel, K. B.; Long, J. R.; McGaughey, A. J. H.; Gomez-Gualdrón, D. A.; Wilmer, C. E. High-throughput screening of hypothetical metal-organic frameworks for thermal conductivity. *npj Computational Materials* **2023**, *9* (1), 11.
- (42) Areán, C. O.; Chavan, S.; Cabello, C. P.; Garrone, E.; Palomino, G. T. Thermodynamics of hydrogen adsorption on metal-organic frameworks. *ChemPhysChem* **2010**, *11* (15), 3237–3242.

(43) Kapelewski, M. T.; Geier, S. J.; Hudson, M. R.; Stück, D.; Mason, J. A.; Nelson, J. N.; Xiao, D. J.; Hulvey, Z.; Gilmour, E.; FitzGerald, S. A.; et al. M_2 (m-dobdc) ($M = \text{Mg, Mn, Fe, Co, Ni}$) metal-organic frameworks exhibiting increased charge density and enhanced H_2 binding at the open metal sites. *J. Am. Chem. Soc.* **2014**, *136* (34), 12119–12129.

(44) Parks, G.; Boyd, R.; Cornish, J.; Remick, R. *Hydrogen station compression, storage, and dispensing technical status and costs: Systems integration*; National Renewable Energy Lab (NREL), Golden, CO, 2014.